Nuclear Model Calculations on the Excitation Functions of Some Radionuclides Produced by Proton Cyclotron

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Abstract: By using the intermediate energetic proton induced reactions, we can produce radionuclides and these radionuclides can be used in medicine and industry. In the last decade, a big success has been provided usage of radionuclides. Nuclear reaction calculations which are based on standard nuclear reaction models can be helpful for determining the accuracy of various parameters of nuclear models and experimental measurements. In this study, production routes of medical isotopes used for diagnostic or a therapeutic radionuclide such as ^{225}Ac, ^{140}Nd, ^{43}Sc and ^{44}Ti were investigated in a range of 10–50 MeV incident proton energy. The excitation functions for (p,2n) reactions were calculated by equilibrium and pre-equilibrium reaction mechanisms. The pre-equilibrium calculations were calculated by using hybrid, geometry dependent hybrid and cascade exciton model. The reaction equilibrium component was calculated with a traditional compound nucleus model developed by Weisskopf-Ewing. Calculation results have been also compared with the available measurements in literature.

1. Introduction

Nuclear reactions induced by incident intermediate and high energetic protons are very important because of wide range technical applications. Especially, the radioisotopes obtained from using charged particles play an important role in medical applications (Aydın et al. 2008; Beyer 2006; Qaim 2002; Qaim 2001). A medical radioisotope can be classified as a diagnostic or a therapeutic radionuclide, depending on its decay properties. These radionuclides are used in diagnostic studies via emission tomography, i.e. Positron Emission Tomography (PET) and Single Photon Emission Computed Tomography (SPECT), and in endoradiotheraphy (internal therapy with radio nuclides) (Qaim 2001; Wolf & Jones 1983).

Nowadays, by using cyclotrons and nuclear reactors can be produced a lot of radioisotopes (Beyer 2006). A cyclotron can accelerate alpha particles to 28-30 MeV and also it can principally accelerate proton particles to energies higher than 30 MeV. Consequently, higher reaction processes such as (p,4n) or generally (p,xn) or even (p,xn,yp) processes are possible (x = 1,2,3… and y = 1,2,3, …). Such a multipurpose cyclotron with the option of high particle beam intensity and well developed tools for beam diagnosis and a certain variation of particle beam energy is an universal instrument supporting commercial isotope production and Research and Development (R&D) in the field of medical isotope application for diagnosis and therapy. Especially (R&D) needed for development of alternative technologies producing carrier-free radioisotope preparations for therapy.

Recently, many evaluated excitation functions of commonly used production reactions can be found in the literature (Broeders & Konobeyev 2007; Sohn & Mattes 1992; Broeders et al. 2006; Gul 2001). Nuclear reaction calculations which are based on standard nuclear reaction models can be helpful for determining the accuracy of various parameters of nuclear models and experimental measurements. In this study, the new calculations on the excitation functions of ^{226}Ra(p,2n)^{225}Ac, ^{141}Pr(p,2n)^{140}Nd, ^{44}Ca(p,2n)^{43}Sc and ^{45}Sc(p,2n)^{44}Ti reactions have been carried out up to 50 MeV incident proton energy. In these calculations, the pre-equilibrium and equilibrium effects have been investigated. The pre-equilibrium calculations involve the geometry dependent hybrid model, hybrid model and the cascade exciton model. Equilibrium effect has been calculated according to the Weisskopf–Ewing model. The calculated results have been compared with the experimental data taken from the literature.

2. Calculation Methods and Parameters Used in Nuclear Reaction Models

The Cascade-Exciton Model (CEM) assumes that the nuclear reactions proceed through three stages: INC, pre-equilibrium and equilibrium (or compound nucleus). Generally, these three components may contribute to any experimentally measured quantity in particular, for the inclusive particle spectrum (Gudima et al. 1983; Barashenkov & Toneev 1972; Barashenkov et al. 1969), we have

$$\sigma(p)\,dp = \sigma_{in}\left[N^{cas}(p) + N^{prq}(p) + N^{eq}(p)\right]dp, \tag{1}$$

where p is a linear momentum, N^{cas} , N^{prq} and N^{eq} are the cascade , the pre-equilibrium and the equilibrium components, respectively. The inelastic cross section σ_{in} is not taken from the experimental data or independent optical model calculations, but it is calculated within the cascade model itself. Hence the CEM predicts the absolute values for calculated characteristics and does not require any additional data or special normalization of its results.

The INC calculations results indicated that the exciton model gave only a prescription for calculating the shape of the pre-equilibrium spectrum and the exciton model deficiency resulted from a failure to properly reproduce enhanced emission from the nuclear surface (Harp & Miller 1971; Feshbach et al. 1980; Tamura et al. 1982). In order to provide a first order correction for this deficiency the hybrid model was reformulated by Blann (Blann 1971; Blann 1975; Blann & Bisplinghoff 1982; Blann et al. 1976). This model, known as geometry dependent hybrid model (GDH) has been developed considered as density distribution of nuclei by Blann and Vonach (Blann & Vonach 1983).

In the density dependent version, the GDH takes into account the density distribution of the nucleus (Blann et al. 1976; Blann & Vonach 1983). This means a longer mean free path at the surface of the nucleus because of a lower density, and a limit to the depth of the holes below the Fermi energy. The differential emission spectrum is given in the GDH as

$$\frac{d\sigma_v(\varepsilon)}{d\varepsilon} = \pi \lambda\!\!\!^{-2} \sum_{\ell=0}^{\infty}(2\ell+1)T_\ell P_v(\ell,\varepsilon), \tag{2}$$

where $\lambda\!\!\!^{-}$ is the reduced de Broglie wavelength of the projectile and T_l represents the transmission coefficient for the lth partial wave. $P_v(\ell,\varepsilon)$ is number of particles of the type v (neutrons and protons) emitted into the unbound continuum with channel energy between ε and $\varepsilon + d\varepsilon$ for the lth partial wave. The GDH model is made according to incoming orbital angular momentum l in order to account for the effects of the nuclear-density distribution. This leads to increased emission from the surface region of the nucleus, and thus to increased emission of high-energetic particles. In this way the diffuse surface properties sampled by the higher impact parameters were crudely incorporated into the pre-compound decay formalism in the GDH.

3. Results and discussion

In the calculations of the hybrid and GDH model, the code as ALICE/ASH was used. The ALICE/ASH code is an advanced and modified version of the ALICE codes (Broeders et al. 2006). The generalized superfluid (Ignatyuk et al. 1979) has been applied for nuclear level density calculations in the ALICE/ASH code. The ALICE-91 (Blann 1991) and ALICE/ASH codes use the initial exciton number as n_0 =3. But in these models the different neutron (n) and proton (p) exciton numbers are used in the pre-equilibrium GDH model calculations. In details, the other code model parameters can be found in Ref. (Broeders et al. 2006). In the present work,

Cascade Exciton Model (CEM) calculations have been made by using CEM95 (Mashnik 1995) (extended version of the previous version named CEM92M (Gudima et al. 1983; Mashnik & Toneev 1974) computer code with the level density parameter by using the systematic of Iljinov et al. (Iljinov et al. 1992). In details, the other code model parameters can be found in Ref. (Mashnik 1995; Mashnik & Toneev 1974).

Although there are some discrepancies between the calculations and the experimental data, in generally, hybrid and GDH model calculations (with ALICE/ASH) are in best agreement with the experimental data above 10 MeV incident proton energies in Figs. 1-3 except for Fig. 4. While the Weisskopf-Ewing model (equilibrium-ALICE/ASH) calculations are only in agreement with the measurements up to 15-30 MeV energy regions (in

Figs.1,2), the cascade exciton model calculations and hybrid model calculations are in good harmony with the experimental data above 10 MeV incident proton energies except for Figs.1,3. As a result, the production of ^{225}Ac, ^{140}Nd, ^{43}Sc and ^{44}Ti radionuclides can be employed at a medium-sized cyclotron.

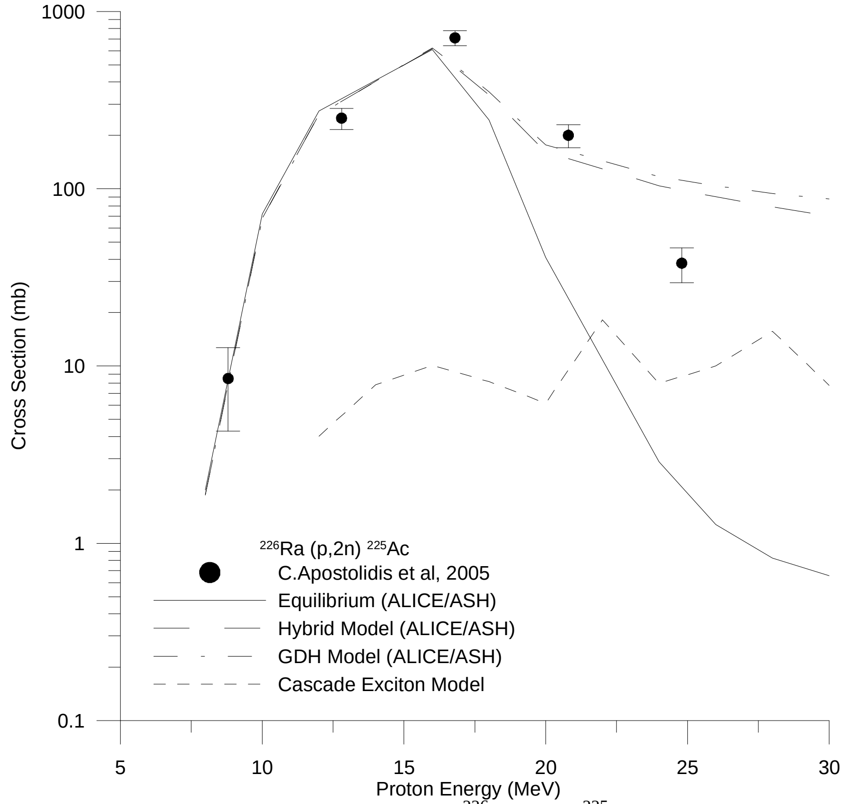

Fig. 1. The comparison of calculated excitation function of ^{226}Ra(p,2n)^{225}Ac reaction with the values reported in Ref. (EXFOR/CSISRS 2007).

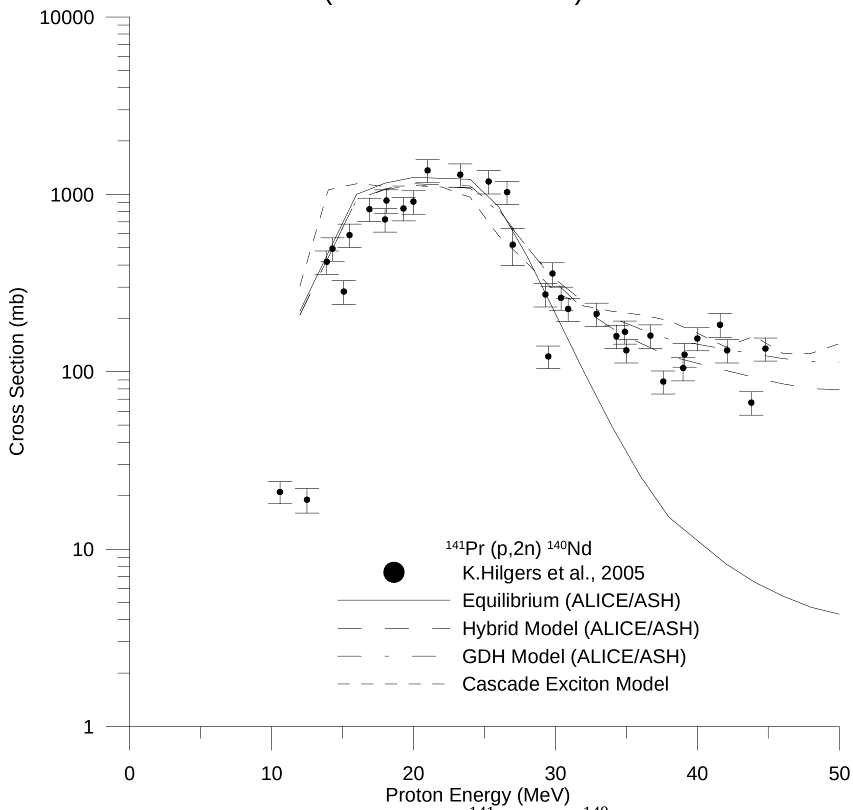

Fig. 2. The comparison of calculated excitation function of ^{141}Pr(p,2n)^{140}Nd reaction with the values reported in Ref. (EXFOR/CSISRS 2007).

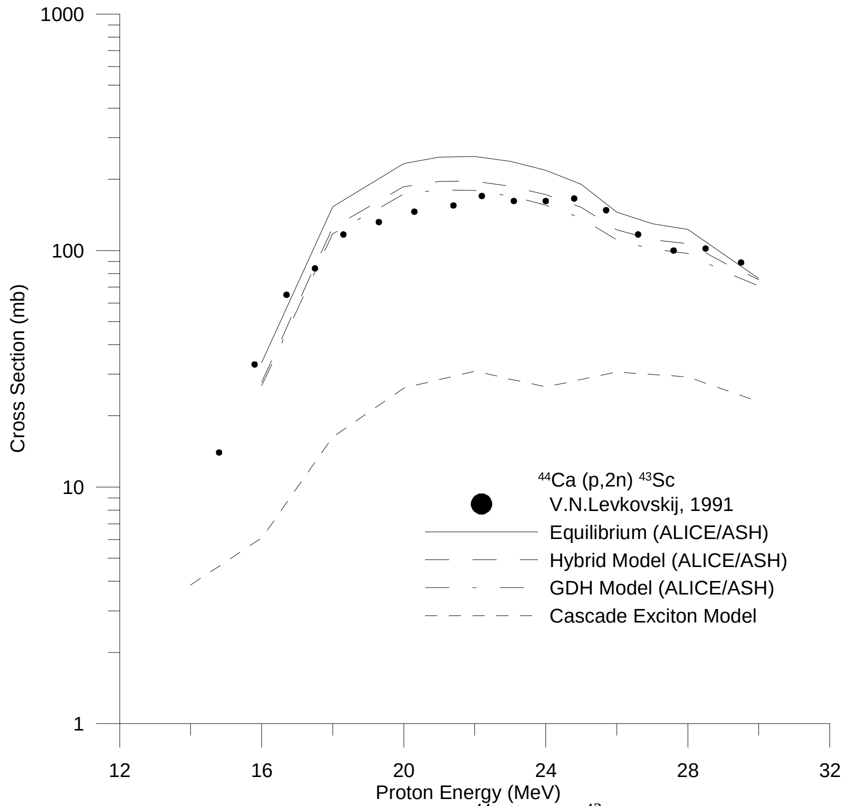

Fig. 3. The comparison of calculated excitation function of ^{44}Ca(p,2n)^{43}Sc reaction with the values reported in Ref. (EXFOR/CSISRS 2007).

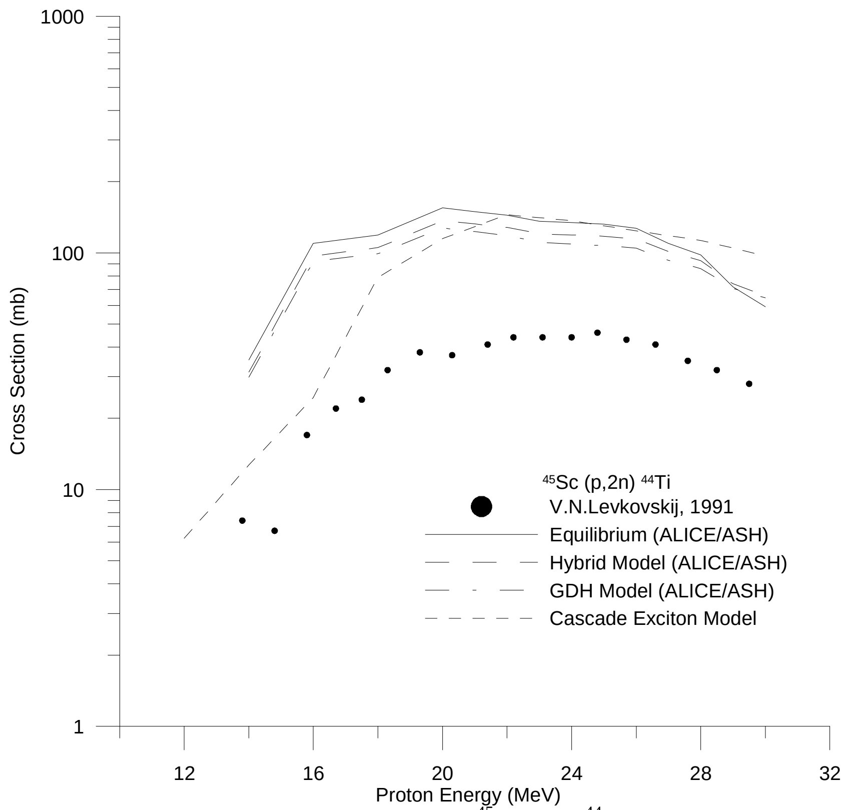

Fig. 4. The comparison of calculated excitation function of ^{45}Sc(p,2n)^{44}Ti reaction with the values reported in Ref. (EXFOR/CSISRS 2007).

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